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SPRAY DRIED FERRITE POWDERS FOR ARC PLASMA SPRAYING.(U)
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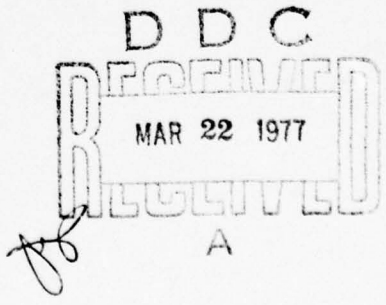
SPRAY DRIED FERRITE POWDERS FOR ARC PLASMA SPRAYING

Janina J. Mitchell

Electronics Technology & Devices Laboratory

February 1977



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The physical characteristics of spray dried lithium ferrite powders to be used in the arc plasma spraying process are discussed. The effects of spray dry parameter changes on the flow characteristics, average size and size distribution of the spherical agglomerates produced by this process, are established. Also established are the relationships between these spray dried lithium ferrite powder characteristics and the magnetic properties of the bulk lithium ferrite produced by arc plasma spraying.

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1. INTRODUCTION

The technique of arc plasma spraying phase shifter elements, as developed by R. W. Babbitt[1], consists of depositing a molten ferrite powder onto a rotating, preheated dielectric substrate. Ideally, a fast and dense deposit occurs when all the powder impinging on the target is in a molten state. However, there are intrinsic limitations as to the maximum particle size and particle size distribution of a powder that can be successfully melted by the arc plasma gun.[2] This makes the powder characteristics a significant parameter of the arc plasma spray (APS) process. Though suitable phase shifter elements had been obtained by arc plasma spraying commercially available spray dried powders, which were prepared for conventional pressing and sintering, it was anticipated that ferrite deposits with higher densities and better reproducibilities would result if the particle size and size distribution of the spray dried powders were optimized for arc plasma spraying.

2. PRELIMINARY EXPERIMENTATION

To determine the powder characteristics required for arc plasma spraying, the following investigation was made:

a. Procedures A completely reacted lithium ferrite powder, developed for S-band phase shifter elements, with a $4\pi M_s$ of 600 gauss, was used. This powder, which had been spray dried, was separated by mechanical sieving into four batches composed of the following particle size* ranges: 4 to 53 μm, 53 to 75 μm, 4 to 75 μm and 75 to 210 μm. Additional sieving with a sonic sifter was required to remove the fine particles still remaining in Batch IV(75 to 210 μm). These known particle size batches were individually arc plasma sprayed into $2\frac{1}{2}$ inch long phase shifter elements. To obtain the 1/8 inch thick walls of the elongated ferrite toroid enclosing the dielectric as required for an S-band phase shifter element (Figure 1), it was necessary to arc plasma spray approximately $\frac{1}{4}$ inch of ferrite around a rotating dielectric-substrate before machining to the desired dimensions. The APS parameters were:**

arc current 330 - 350 A
working distance 2 3/4 ±1/8 inches
arc gas(argon/helium) 75-80/3 cubic feet per hour(cf/hr)
carrier gas(oxygen) 50 - 60 cf/hr
powder feed 80 cf/hr

*In this study, particle size is defined as the spherical agglomerate obtained after spray drying.
**APS parameters varied for Batch II

1. Richard W. Babbitt, "Arc Plasma Sprayed C-Band Lithium Ferrite Phase Shifters", IEEE Transactions on Magnetics, vol. Mag-11, No. 5, September 1975, pp 1253-1255.
2. Richard W. Babbitt, "Arc Plasma Fabrication of Ferrite-Dielectric Composites," American Ceramics Society Bulletin, Vol. 55, No. 6, June 1976.

Figure 1 - S-Band Phase Shifter Element

The densities of the APS samples were obtained by using the American Society for Testing Materials (ASTM) procedure C376-56. The magnetic measurements were made on toroids cut from the ferrite portion of the phase shifter element, then measured in accordance with ASTM Special Technical Publication #371.

b. Results: Figure 2 is a comparison of the bulk densities of the ferrite samples obtained from the four batches of powder which were arc plasma sprayed. The vertical lines are composed of data points showing the individual sample densities obtained for each batch of powder.

The percentage figures indicate the average percent density obtained for each batch of powder as compared with the control sample which had a bulk density of 4.1806. The control sample had been fabricated by ceramic techniques which are used conventionally to obtain densities greater than 98% of the theoretical density. The best average percent density for the APS samples is 95.5% and is from the first batch which contains the smallest particle size powder. Even though the number of samples studied were limited, some definite trends are shown on this graph. The slope of the broken line indicates that the average bulk density is inversely proportional to the particle size, i.e. the smaller the particle size, the higher the density. Comparison of the lengths of the vertical lines enclosing the individual density data points of Batches I and III indicate that the bulk density spread is directly proportional to the particle size range. Specifically, the larger the particle size distribution, the greater the bulk density variation.

The two results obtained from Batch II (53-75 μm size particles) are included for comparative purposes only, as the APS parameters used for this batch were modified in order to obtain a faster deposition rate. Probably the density spread would be larger if more samples were tested, and the average density higher if the arc plasma parameters were identical with those used on the other batches of powder.

The results obtained from Batch IV (75 to 210 μm size particles) appear to be an anomaly. But a review of the APS parameters of these samples showed that approximately $2\frac{1}{2}$ times more powder (by weight) and twice the spray time was required to produce samples from this batch of powder. It is believed that this increased consumption in time and powder was caused by an incomplete melt of these large spheres (that portion of a particle which is not in a molten state will not adhere to the rotating core).

Table 1 compares the averages of the densities with the remanences (B_r)* and squareness ratios (B_r/B_m) of the conventional standard and the four batches of powders. The maximum variation from the average in both density and remanence was $\pm 7\%$, which occurred in the samples obtained from Batch III (4-75 μm size). The remanences and squareness ratios listed are the results

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*A high remanence is desirable for phase shifter applications because it will produce a higher differential phase shift.

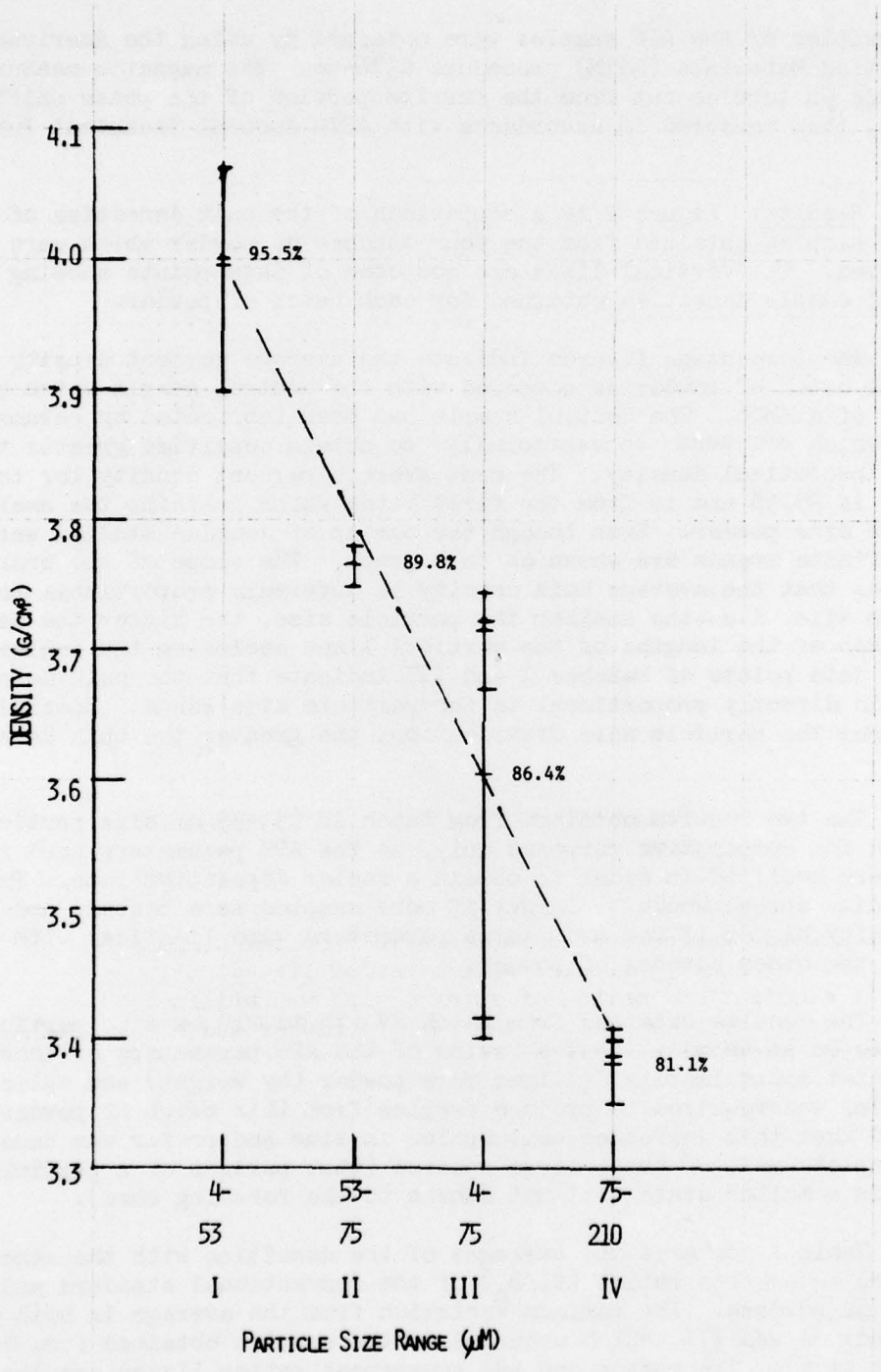

Figure 2 - Arc Plasma Sprayed Ferrite Density vs. Powder Particle Size

of measurements made at five times the coercive force. An examination of the data on this table shows that the remanence and the squareness ratio of these arc plasma sprayed ferrites are directly proportional to their density which is inversely proportional to the particle size of the starting material.

TABLE 1. DENSITY VS. REMANENCE (B_r) AND B_r/B_m*

PARTICLE SIZE (μm)	DENSITY (g/cm^3)	B_r (Gauss)	B_r/B_m
Conventional Standard ($4\pi M_s$ = 600G)	4.1806	372	0.942
4-53	3.8955	370.4	0.870
53-75	3.7655	352.9	0.854
4-75	3.6764	329.9	0.844
75-210	3.3979	262.4	0.784

As a result of these studies it was decided to optimize the spray drying procedure used to prepare ferrite powders for arc plasma spraying. The desired objective was to produce a ferrite powder with good flow properties composed of small spheres having a narrow size distribution.

3. SPRAY DRYING PROCEDURE

A 750 gauss lithium ferrite powder was used for the spray drying experimentation which was conducted in the Niro Atomizer Company plant. Because of the desire for small particles with a narrow size distribution, their centrifugal atomization[3] method of spray drying was utilized. In this method the aqueous slurry composed of the ferrite powder and binders is fed around the central shaft of a rotating disc to be discharged at high speed from the periphery of the disc into the drying chamber which contains the continually flowing, hot-drying air. The procedure is essentially the same for both their small laboratory dryer and the large utility spray dryer, both of which were used for these experiments. However, the laboratory model has only one powder collection point which is at the base of the small cyclone; whereas the utility model produces a chamber fraction collected from the base of the chamber and the cyclone fraction.

The spray drying parameters, varied during experimentation, are

*Maximum flux at five times coercive force.

3. David A. Lee, "Comparison of Centrifugal vs. Nozzle Atomization in Spray Dryer Operations," presented at the White Ware Division of the American Ceramic Society at the Annual Meeting, 2 May 1973.

given in Table 2.

TABLE 2. PARAMETERS VARIED DURING SPRAY DRYING

PARAMETERS	SPRAY DRYER MODEL	
	LABORATORY	UTILITY
Inlet Air Temp °C	125 - 180	160 - 230
Outlet Air Temp °C	90 - 110	90 - 110
Wheel Speed (r/min x 10^3)	25 - 40	18 - 22.3
% Feed Solids	50 - 55	55 - 62
Feed Rate (lb/min)	3.75 - 6.25	20 - 69
Type Binder Used	2 - 5% PEG* 2% PVA** 3% PVA + 1% PEG***	5% PEG* 2 - 3% PVA** 2% PVA + 1% PEG***

The differences in inlet air temperature, wheel speed and feed rates are a function of the model sizes. The percentage of feed solids was the amount of ferrite powder plus binder added to the solution. The binders were polyvinyl alcohol(PVA) and/or polyethylene glycol (PEG). When PEG was used alone it was carbowax 6000 but when combined with the PVA it was polyethylene glycol 200. The PVA used was Gelvatol 20-30.

4. RESULTS AND DISCUSSION

After the completion of a spray dry run, the powder was examined for flow properties, residual moisture content and particle size distribution. These rapid examinations were used to determine the spray dry parameter changes. However, the final test for a powder was how it flowed through the arc plasma gun. For example, those spray dried powders in which carbowax 6000 was used as the binder were quickly eliminated because of their relatively poor flow characteristics and the binder's low melting point which caused the external feed line of the arc plasma gun to become clogged.

a. Best Spray Dry Parameters**** Table 3 shows the spray dry parameters of three runs which produced powders with good flow properties. Both inlet and outlet air temperature, feed rate and percentage feed solids are higher for the utility spray dryer. This is due to its larger size and the incorporation of design features for fast spray drying of large batches.

*PEG = Carbowax 6000
**PVA = Gelvatol 20-30
***PEG = Polyethylene Glycol 200
****The parameters listed are for the specific spray dryers used. Slight modifications may be necessary to obtain the same results from different spray dryers of the same type.

The wheel speed, as listed for the centrifugal disc of the laboratory run, seems greater; but if it were expressed as the peripheral speed of the edge it would be 8560 ft/min for the 40,000 r/min of the laboratory model and 8660 ft/min for the 22,000 r/min of the utility model. At this speed the difference is negligible. The binders varied for each run as designated.

TABLE 3. SPRAY DRY PARAMETERS OF THREE RUNS

PARAMETERS	SPRAY DRYER MODEL		
	LABORATORY	UTILITY	
	I	II	III
Inlet Air Temp ^{o}C	125	230	230
Outlet Air Temp ^{o}C	95	105	105
Wheel Speed (r/min x 10^3)	40	21.9	22.05
% Feed Solids	50	60	55
Feed Rate (lb/min)	6.25	69	53.2
Type Binder Used	2% PVA**	2% PVA + 1% PEG***	3% PVA**

b. Particle Size Comparisons. The powders obtained from these three runs were analyzed for particle size distribution using a sonic sifter. The average particle size was determined by scanning electron microscope (SEM) count.

The results of the sonic sieve analyses of the spray dried powders obtained from these runs are listed in Table 4. The chamber fractions of the utility runs are identified by the letter A and the cyclone fractions by the letter B.

**PVA = Gelvatol 20-30
***PEG = Polyethylene Glycol 200

TABLE 4. SONIC SIEVE ANALYSIS OF THE THREE SPRAY DRY RUNS

US Standard Sieve Number	Sieve Opening (μm)	Spray Dry Runs (Weight % Retained on Sieve) I	*II-A	**II-B	III-A	III-B
100	150	0.9	0.1	0.3	1.5	8.8
200	75	0.6	0.3	1.6	3.1	10.3
230	63	0.2	0.4	2.1	1.2	8.5
270	53	0.4	2.6	3.9	4.5	9.5
325	45	1.5	21.7	17.5	22.2	10.7
400	37	5.3	31.7	13.7	31.4	20.3
Collector	< 37	91.2	43.3	60.9	36.0	32.0

An examination of Table 4 shows that the spray dried powder produced by Run I is the best of these three runs for arc plasma spraying. With 98% by weight of the powder being finer than 53 μm in size, Run I has the smallest particle size range. Additionally, this powder has the smallest average particle size because 91.2% by weight is finer than 37 μm and the minimum particle size (SEM) is 2.75 μm.

Figure 3 is a bar graph of these three runs showing the distribution of 91% by weight of the smallest particles in each batch. Knowing that the Run I powder is the best of the three dry runs, it is expected that the particle size range would be greater for the other batches. But the greater particle size range in the cyclone portion of the runs, especially Run III-B, was contrary to the expectation that the cyclone portions would have smaller particles. Since these results had been duplicated in other sieve analyses, the possibility that the ultrasonic vibrations were causing the minute spheres to reform into larger spheres created some doubt about the validity of this type of analysis. However, SEM photographs taken to determine average particle size by count disproved this theory.

Figure 4 is a SEM photograph of the Batch III-B powder magnified 720 times. The average particle size was determined to be 26 microns. The lack of clarity or fuzziness of the spherical outlines in this picture is caused by the unformed particle ferrite material present in this powder sample.

Figure 5 is a SEM photograph of Run I magnified 720 times. The average particle size by count is 20 microns which is smaller than the 26

*II-A = Powder of Chamber Fraction of Utility Run II
**II-B = Powder of Cyclone Fraction of Utility Run II

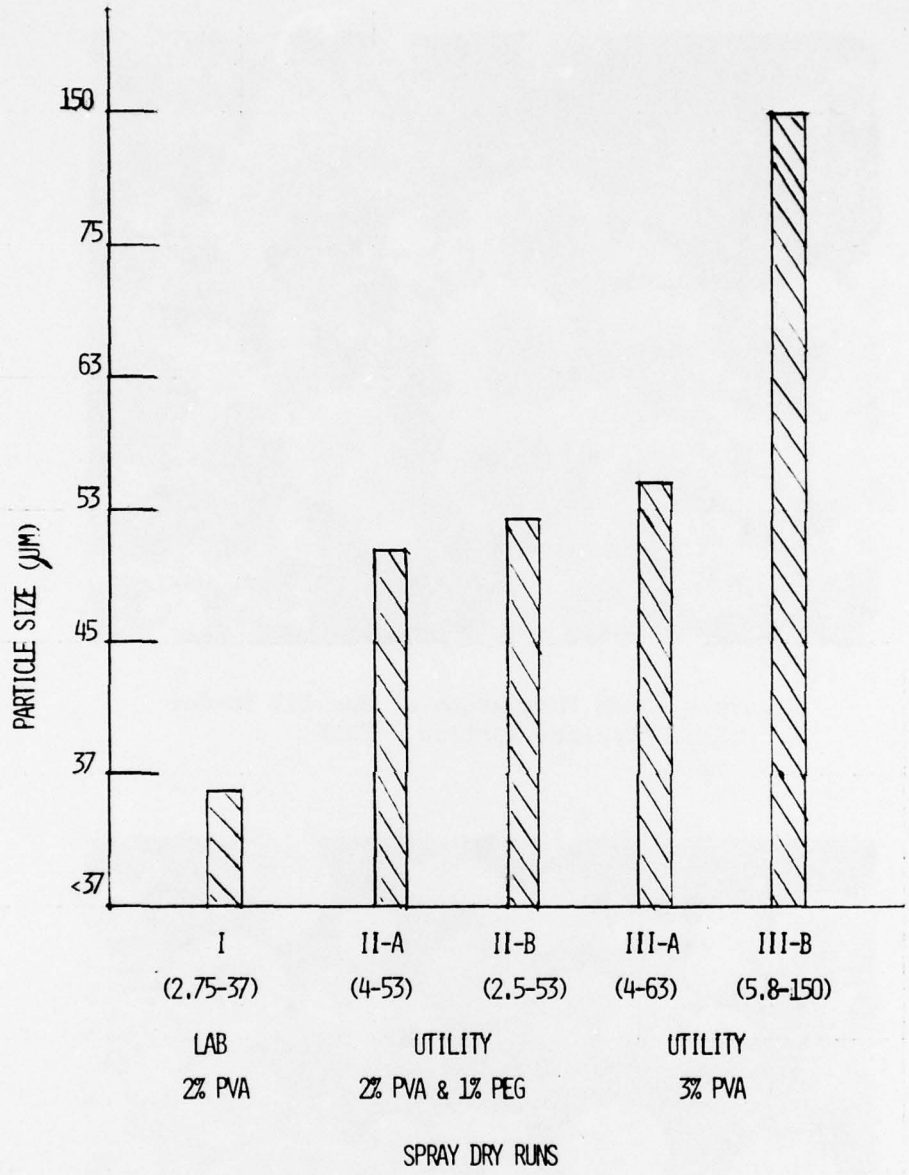

Figure 3 - Particle Size Distribution of Three Spray Dry Runs

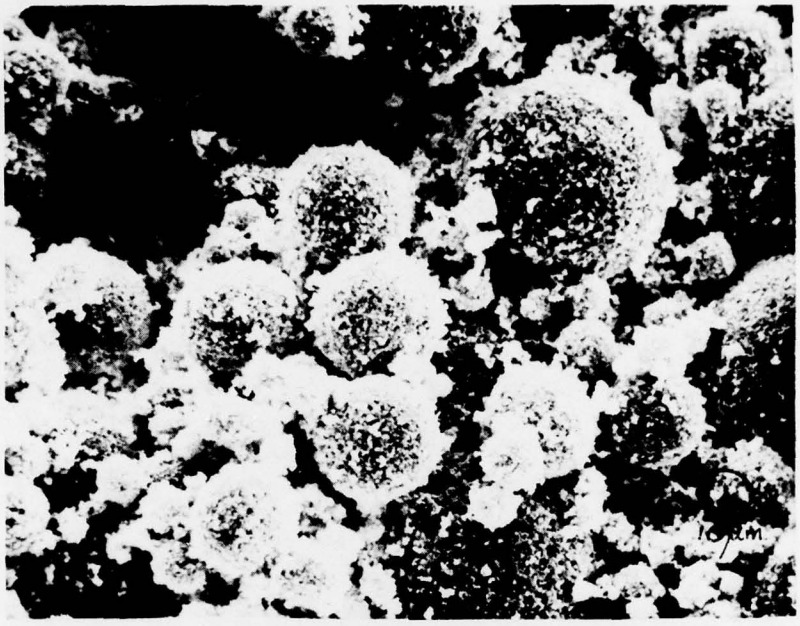

Figure 4 - SEM Photograph of Run III Powder
Cyclone Portion 720X

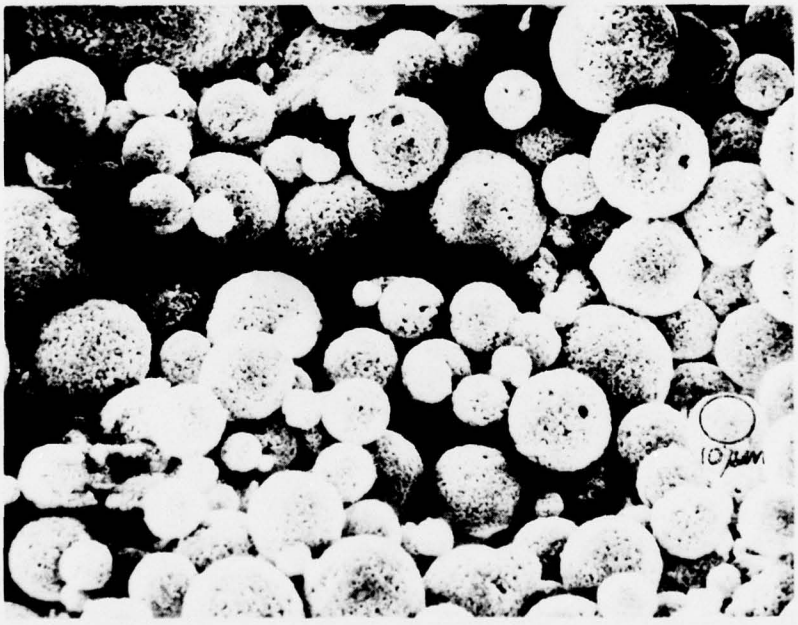

Figure 5 - SEM Photograph of Run I
Powder 720X

microns of the previously discussed powder, yet the spherical particle outlines are clear and distinct. Therefore, it is not the very small particle size that makes the sieve analysis results seem unreliable but the unformed spheres of stray material which the ultrasonic vibrations cause to fly around until they clog the openings of the sieves.

c. Densities of APS powders Runs I and III* of these spray dried powders were individually arc plasma sprayed into $2\frac{1}{2}$ inch long phase shifter elements. The APS parameters were:

Arc Current	300A
Working Distance	2 3/8 in $\pm$1/8 in
Arc Gas (Argon/Helium)	85/3 cf/hr
Carrier Gas (O_2)	80 cf/hr
Powder Feed	80-85 cf/hr

The vertical lines in Figure 6 compare the densities and density spreads of these arc plasma sprayed ferrite samples. The wider horizontal line on each of the vertical lines, which enclose the density data points, is the average density of the batch. The powder characteristics are given in the box on the right of the figure.

The differences in the density spreads of the APS samples obtained from these three batches or powders are directly related to their respective particle size distribution ratios. These ratios, which provide a means for a quantitative comparison between the various particle size distributions, and the resultant density spreads shown in Figure 6, are obtained by dividing the largest particle size by the smallest one of the particle size ranges, given in Figure 3. The computed ratios are 13.5 for Run I, 15.8 for Run III-A and 25.9 for Run III-B. Comparisons between these ratios with their bulk density spreads (Figure 6) confirm the preliminary experimentation result that the bulk density spread is directly proportional to the particle size range.

Consistent with the results of Figure 2, powder I with an average particle size (SEM) of 20 μm has a higher density than powder III-A whose average particle size is 40. Yet, powder III-A has a higher average density than III-B which has an average particle size of 26 μm as obtained by electron microscope count. However, if the weighted averages of the sieve analysis results of these three powders are computed, the following average sizes for the powders are obtained: 22.2 μm for Run I, 39.6 μm for Run III-A and 55.6 μm for Run III-B. Comparisons of these computed average sizes with their average densities confirm both the previously developed premise that the smaller the particle size, the higher the resultant APS density, and the validity of the ultrasonic sieve analysis results for the arc plasma spray process.

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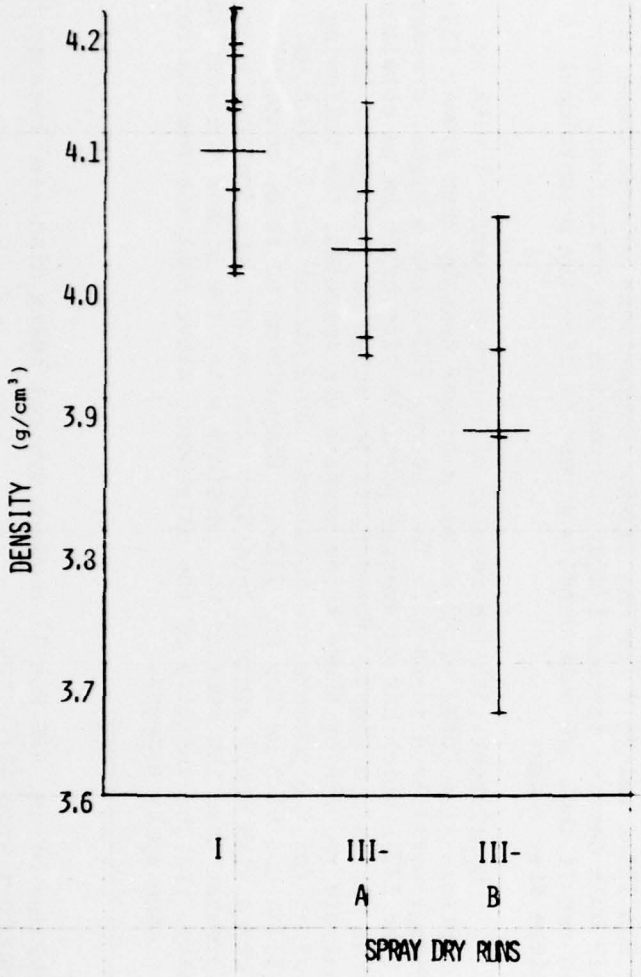
*The densities of the APS Run II powders are not being discussed because the APS parameters were different.



'POWDER CHARACTERISTICS

	I	III-A	III-B
Type Dryer	Lab	U-Ch*	U-Cy*
% PVA	2	3	3
Av. Particle Size(μ)	20	40	26
Sieve Analysis (%)			
<37μ	91.2	36.0	32.0
<53μ	98.0	90	63.0
>75μ	1.4	2.2	19.1

*U-Ch = utility-chamber

U-Cy = utility-cyclone

Figure 6 - Density vs. Spray Dry Runs

The one magnetic property not mentioned has been the $4\pi M_s$ of these ferrites. The $4\pi M_s$ of the APS samples obtained from the 600 gauss powder, used in the preliminary experimentation, varied from 520 to 581 gauss, while those obtained from the 750 gauss powder varied from 710 to 790 gauss. This spread in $4\pi M_s$ is attributed to the density variations of the test samples.

5. CONCLUSIONS

The best parameters established for spray drying a ferrite powder for arc plasma spraying are those of Run I for the laboratory spray dryer. Microscopic examination of this powder showed that it consisted of well shaped, discrete spheres to which its good flow properties are attributed. The average particle size, as determined by SEM count, was 20 microns and, as indicated by the sonic sieve analysis, the particle size spread was the smallest.

The importance of control of the particle size and particle size distribution characteristics of the spray dried ferrite powders, used for the arc plasma spraying process, has been demonstrated. Also, the best means of determining these required powder characteristics has been determined. The control of these powder characteristics eliminates one of the critical variables in the arc plasma spray process, thereby simplifying the determination of the optimum arc plasma parameters for increased production of reliable and reproducible ferrite devices.

However, neither the effect nor the feasibility of producing a spray dried powder consisting of an average particle size, smaller than the 20 μm size, with a narrow size spread, nor the relationship between the minimum particle size and good flow characteristics, has yet been determined.

6. ACKNOWLEDGEMENTS

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